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Haloethers: Ambient Water Quality Criteria

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HALOETHERS

Ambient Water Quality Criteria

Criteria and Standards Division
Office of Water Planning and Standards
U.S. Environmental Protection Agency
Washington, D.C.

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Section 304(a) of the Clean Water Act (33 U.S.C. 1314(a)), requires EPA to publish and periodically update water quality criteria. These criteria are to reflect the latest scientific knowledge on the identifiable effects of pollutants on public health and welfare, aquatic life, and recreation. This report presents water quality criteria for haloethers. It presents concentration criteria for the protection of fresh water and saltwater aquatic life. It presents 'safe' concentrations for humans, and in the case of suspect or proven carcinogens, gives various levels of incremental cancer risk. A section 304(a) water quality criterion is a qualitative or quantitative estimate of the concentration of a water constituent or pollutant in ambient waters which, when not exceeded, will ensure a water quality sufficient to protect a specified water use. Under the Act a criterion is a scientific entity, based solely on data and scientific judgement. It does not reflect consi....

KEYWORDS: *Water quality, *Toxicology, *Halogen organic compounds, *Ethers, *Toxic substances, *Water pollution effects(Humans).

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CRITERION DOCUMENT

HALOETHERS

CRITERIA

Aquatic Life

4-bromophenylphenyl ether

For 4-bromophenylphenyl ether the criterion to protect freshwater aquatic life as derived using the Guidelines is 6.2 µg/l as a 24-hour average and the concentration should not exceed 14 µg/l at any time.

For saltwater aquatic life, no criterion for 4-bromophenylphenyl ether can be derived using the Guidelines, and there are insufficient data to estimate a criterion using other procedures.

Human Health

There are insufficient toxicological data to calculate exposure criteria for the haloethers covered in this document.

Introduction

Haloethers are compounds which contain an ether moiety (R-O-R) and halogen atoms attached to the aryl or alkyl groups. Chloroethers appear to be the most important haloethers used commercially and can be divided into two categories, alpha- and non-alpha-chloroethers (EPA, 1975). Chloromethyl methyl ether (CMME) is the only alpha haloether of commercial significance and is used primarily in the synthesis of strong base ion exchange resins used in water conditioning and for chemical separation processes. However, CMME preparations are usually contaminated with 1 to 8 percent bix(chloromethyl) ether (BCME) which has been demonstrated to be a potent carcinogen.

The beta-chloroethers are widespread environmental contaminants. It has been suggested that they are: produced or may be formed as by-products in sizable quantities, released to and appear to persist in the environment, can pass through drinking water treatment plants, and may be carcinogenic. Bis(2-chloroethyl) ether (BCE) is used as a dewaxing agent for lubricating oils and is a useful solvent for naphthenic components (Fairhall, 1949; Jacobs and Scheflan, 1953; Pollard and Lawson, 1955; Mervart, et al. 1960). BCE has also been used to separate butadiene from butylene (Lurie, 1965). The second major use of bis(2-chloroethyl)-ether is in the textile industry as a cleaning agent, a wetting agent and penetrant in combination with diethylene glycol, sulphonated oils, etc. (Browning, 1953; Jacobs and Scheflan, 1953; Allen, 1956). The compound generally is

a good solvent for tars, fats, waxes, oils, resins and pectins, and will dissolve cellulose esters when used with 10 to 30 percent ethanol (Fife and Reid, 1930).

Bis(2-chloroisopropyl) ether is an excellent solvent and extractant for fats, waxes, and greases. It also finds use as a cleaning and spotting agent as well as an additive to paint and varnish removers (Hake and Rowe, 1963; Lurie, 1965).

The alpha-haloethers are more reactive than beta-haloethers due to the two electronegative atoms (oxygen and halogen) which are bonded to the same carbon (Summers, 1955). This difference in reactivity is evident by the different rates of hydrolysis. The most commercially significant haloethers are the chloroethers. Chlorine substitution on ethers tends to increase their density, boiling point, and odor while decreasing their flammability and altering their solubility properties. The fluorine substituted compounds are much more volatile than their chlorinated analogues (EPA, 1975).

The haloethers exist within a wide range of physical properties. For example, boiling points may range from 43.2°C (2,2,2-trifluroethyl vinly ether) to 310°C (4-bromophenylphenyl ether) (Lurie, 1965). Melting points can range from 103.5°C (chloromethyl methyl ether) to -3°C (chloromethyl phenyl ether) (Hawley, 1971). The haloethers are very soluble in benzene, carbon tetrachloride, and acetone (Scheflan and Jacob, 1953) and miscible in all oils (Lurie, 1965).

Table 1 lists the physical properties of some haloethers.

TABLE 1

Physical Properties of Haloethers

Ether	Structure	Boiling Point (deg.C)	Melting Point (deg.C)	d20/4	Vapor Pressure (mm Hg)	Solubility in water (g/100)
bis(chloromethyl)	$(ClCH_2)_2O$	105	-41.5	1.315		
bis(2-chloroethyl) (Chlorex)	$(ClCH_2CH_2)_2O$	178	-46.7	1.2199	$0.73^{20°C}$	1.07
bis(2-chlorisopropyl)	$(ClCH_2CH)_2O$ CH_3	187.3	-96.8 to 99.8	1.1127	$0.71-0.85^{20°C}$	0.17
bis(2-chloroethoxy) methane	$(ClCH_2CH_2O)_2CH_2$	218.1	-32.8	1.234		
Chloromethyl methyl	$ClCH_2OCH_3$	61	-103.5	1.0605		
2-(2-chloroethoxy)ethyl 2-chloroethyl	$ClCH_2CH_2OCH_2CH_2OCH_2CH_2Cl$					
dichloromethyl methyl	Cl_2CHOCH_3	84.5		1.270		
dichloromethyl chloro-methyl	Cl_2CHOCH_2Cl	129		$1.464^{30°C}$		
bis(dichloromethyl)	$Cl_2CHOCHCl_2$	143		$1.558\ 30°C$		
trichloromethyl methyl	Cl_3COCH_3	106-110.5		1.4391		
trichloromethyl dichloromethyl	$Cl_3COCHCl_2$	159				
1-chloroethyl ethyl	$CH_3CHClOC_2H_5$	28_{57mm}		0.9495		
2-chloroethyl ethyl	$ClCH_2CH_2OC_2H_5$	106		0.9945		
bis(1-chloroethyl)	$(CH_3CHCl)_2O$	113		$1.106^{25°C}$		

TABLE 1 (continued)

Ether	Structure	Boiling Point (deg.C)	Melting Point (deg.C)	d20/4	Vapor Pressure (mm Hg)	Solubility in water (g/10)
bis(2-bromoethyl)	$(BrCH_2CH_2)_2O$	115_{32mm}		$1.8227^{27°C}$		
bis(2-chloropropyl)	$(CH_3CHClCH_2)_2O$	188		1.109		
bis(3-chloropropyl)	$(ClCH_2CH_2CH_2)_2O$	215_{745mm}		$1.140^{20/20}$		
bis (4-chlorobutyl)	$(ClCH_2CH_2CH_2CH_2)_2O$	$84\text{-}86_{5.5°C}$		$1.0691^{25°C}$		
2-chloroethyl vinyl	$ClCH_2CH_2OCH{=}CHH_2$	109	-70	1.0493		0.6
chloromethyl ethyl	$ClCH_2OCH_2CH_3$	84		1.0322^{o}_{4}		
chloromethyl phenyl	$ClCH_2OC_6H_5$	88-90	- 3			
2,2-dichloro-1,1-difluoroethyl methyl (methoxyflurane)	$CHCl_2CF_2OCH_3$	104	-35	1.4262		
2,2,2-trifloroethyl vinyl (fluoroxene or fluoromar)	$CF_3CH_2OCH{=}CH_2$	43.2		1.13	286	0.4

Lurie, 1965; Allen, 1956; Tschamler, 1950; Krentz, 1963; Hake and Roe, 1963

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AQUATIC LIFE TOXICOLOGY*

FRESHWATER ORGANISMS

Introduction

The only toxicity data for haloethers, other than for those compounds discussed in the criterion document for chloroalkyl ethers, are for 4-bromophenylphenyl ether and the bluegill, fathead minnow, and *Daphnia magna*.

Acute Toxicity

The bluegill has been exposed to 4-bromophenylphenyl ether and the unadjusted 96-hour LC50 is 4,940 µg/l (Table 1). When this result is adjusted for test conditions and species sensitivity, a Final Fish Acute Value of 690 µg/l is obtained.

Daphnia magna is more sensitive than the bluegill with an unadjusted 48-hour EC50 of 360 µg/l (Table 2). The Final Invertebrate Acute Value for 4-bromophenylphenyl ether is 14 µg/l and this also is the Final Acute Value.

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*The reader is referred to the Guidelines for Deriving Water Quality Criteria for the Protection of Aquatic Life [43 FR 21506 (May 18, 1978) and 43 FR 29028 (July 5, 1978)] in order to better understand the following discussion and recommendation. The following tables contain the appropriate data that were found in the literature, and at the bottom of each table for the calculations for deriving various measures of toxicity as described in the Guidelines.

Chronic Toxicity

A chronic value for 4-bromophenylphenyl ether, 61 ug/l, is derived from an embryo-larval test with the fathead minnow in which adverse effects on survival and growth were observed (Table 3) (U.S. EPA, 1978). After division by the species sensitivity factor (6.7), a Final Fish Chronic Value of 9.1 ug/l is derived. Since no other chronic value for an invertebrate or plant species or a Residue Limited Toxicant Concentration is available, 9.1 ug/l is also the Final Chronic Value.

CRITERION FORMULATION

Freshwater-Aquatic Life

Summary of Available Data

The concentrations below have been rounded to two significant figures.

4-bromophenylphenyl ether

Final Fish Acute Value = 690 µg/l

Final Invertebrate Acute Value = 14 µg/l

Final Acute Value = 14 µg/l

Final Fish Chronic Value = 9.1 µg/l

Final Invertebrate Chronic Value = not available

Final Plant Value = not available

Residue Limited Toxicant Concentration = not available

Final Chronic Value = 9.1 µg/l

0.44 x Final Acute Value = 6.2 µg/l

The maximum concentration of 4-bromophenylphenyl ether is the Final Acute Value of 14 µg/l and the 24-hour average concentration is 0.44 times the Final Acute Value. No important adverse effects on freshwater aquatic organisms have been reported to be caused by concentrations lower than the 24-hour average concentration.

CRITERION: For 4-bromophenylphenyl ether the criterion to protect freshwater aquatic life as derived using the Guidelines is 6.2 µg/l as a 24-hour average and the concentration should not exceed 14 µg/l at any time.

Table 1. Freshwater fish acute values for haloethers (U.S. EPA, 1978)

Organism	Bioassay Method*	Test Conc.**	Chemical Description	Time (hrs)	LC50 (ug/l)	Adjusted LC50 (ug/l)
Bluegill, Lepomis macrochirus	S	U	4-Bromophenyl-phenyl ether	96	4,940	2,700

* S = static

** U = unmeasured

Geometric mean of adjusted values: 4-Bromophenylphenyl ether = 2,700 µg/l $\frac{2,700}{3.9}$ = 690 µg/l

Table 2. Freshwater invertebrate acute values for haloethers (U.S. EPA, 1978)

Organism	Bioassay Method*	Test Conc.**	Chemical Description	Time (hrs)	LC50 (ug/l)	Adjusted LC50 (ug/l)
Cladoceran, *Daphnia magna*	S	U	4-Bromophenyl-phenyl ether	48	360	300

* S = static

** U = unmeasured

Geometric mean of adjusted values: 4-Bromophenylphenyl ether = 300 µg/l $\frac{300}{21} = 14$ µg/l

Table 3. Freshwater fish chronic values for haloethers (U.S. EPA, 1978)

Organism	Test*	Limits (ug/l)	Chronic Value (ug/l)
4-Bromophenylphenyl ether			
Fathead minnow, Pimephales promelas	E-L	89-167	61

* E-L = embryo-larva

Geometric mean of chronic values = 61 µg/l $\quad \frac{61}{6.7} = 9.1$ µg/l

Lowest chronic value = 61 µg/l

CRITERION FORMULATION

Saltwater-Aquatic Life

Summary of Available Data

No appropriate data are available for saltwater organisms and any haloether other than those discussed in the criterion document for chloroalkyl ethers.

CRITERION: No saltwater criterion can be derived for any haloether using the Guidelines because no Final Chronic Value for either fish or invertebrate species or a good substitute for either value is available, and there are insufficient data to estimate a criterion using other procedures.

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Mammalian Toxicology and Human Health Effects

I. Introduction

The EPA is currently charged with establishing water quality criteria for halogenated ethers. This document covers chlorinated aromatic ethers including:

chlorophenyl phenyl ethers
bromophenyl phenyl ethers
polychlorinated diphenyl ethers

A separate document in this series, entitled "Chloroalkyl ethers," covers the following compounds:

bis(chloromethyl)ether
bis(2-chloroethyl)ether
2-chloroethyl vinyl ether
bis(2-chloroisopropyl)ether
bis(2-chloroethoxy)methane

II. Exposure

A. Ingestion

1. Water

Qualitative identifications of several haloethers in raw and finished water have been reported. This information is summarized in Table 1. The nomenclature used in specifying some haloethers creates a certain amount of confusion in evaluating these monitoring data. For instance, in Table 1, pentachlorophenoxy methyl ether is probably the same as pentachlorophenyl methyl ether. The names used in Table 1 are those given in the various references cited.

Shackelford and Keith (1976) have compiled information on the frequency of organic compounds identified in water. This information was taken from both the published literature and unpublished results of survey analyses from EPA Regional Laboratories and Research Laboratories. Although actual levels of the haloethers in waters are not specified, a breakdown is given of the various types of waters found to be contaminated. This information is presented in Table 2.

The study by Ewing and coworkers (1977) is based on the analysis of 204 water samples collected from fourteen heavily industrialized river basins. As indicated in Table 1, pentachlorophenyl methyl ether was found in 12 samples (5.88%), with other chlorophenyl ethers found less frequently. As indicated above, the reports of "pentachlorophenoxy methyl ether" probably refer to pentachlorophenyl methyl ether. The studies by Friloux (1971) and the U.S. Environmental Protection Agency (1972) both summarized in Table 1, were conducted in the New Orleans area.

Table 1. Haloethers Qualitatively Identified in Water
(see text for details)

Haloether \ Reference	Shackelford and Keith, 1976*	Ewing et al., 1977*	Friloux, 1971	U.S. EPA, 1972
Bromophenyl phenyl ether	5		X	X
Bis(4-chlorophenyl)ether	2			
Dichlorophenyl chlorophenyl ether	2			
2,4,4'-Trichloro-2'-hydroxy-diphenyl ether	1			
Dichlorophenyl methyl ether		1		
Trichlorophenyl methyl ether		5		
Tetrachlorophenyl methyl ether		1		
Pentachlorophenyl methyl ether	10	12		
Pentachlorophenoxy methyl ether		2		

*Frequency of occurrence.

Table 2. Frequency Haloethers Identified in Various Types of Water (Shackelford and Keith, 1976)

	Water Type Contaminated*					
				Effluent from:		
	FDW	River	Raw Water	CHEM	RS	STP
Bromophenyl phenyl ether	3	2				
Bis(4-chlorophenyl)ether				2		
Dichlorophenyl chlorophenyl ether				2		
2,4,4'-Trichloro-2'-hydroxy-diphenyl ether				1		
Pentachlorophenyl methyl ether	3	3	1		1	2

*Key

FDW = Finished drinking water
CHEM = Chemical plant
RS = Raw sewage
STP = Sewage treatment plant

In the 1975 National Organics Reconnaissance Survey by the U.S. Environmental Protection Agency, no haloethers were found in the waters of Miami, Florida; Seattle, Washington; Ottumwa, Iowa; or Cincinnati, Ohio (U.S. EPA, 1975).

2. Food

No monitoring data have been found on the levels of haloethers in food.

A bioconcentration factor (BCF) relates the concentration of a chemical in water to the concentration in aquatic organisms, but BCF's are not available for the edible portions of all four major groups of aquatic organisms consumed in the United States. Since data indicate that the BCF for lipid-soluble compounds is proportional to percent lipids, BCF's can be adjusted to edible portions using data on percent lipids and the amounts of various species consumed by Americans. A recent survey on fish and shellfish consumption in the United States (Cordle et al., 1978) found that the per capita consumption is 18.7 g/day. From the data on the nineteen major species identified in the survey and data on the fat content of the edible portion of these species (Sidwell, et al., 1974), the relative consumption of the four major groups and the weighted average percent lipids for each group can be calculated:

Group	Consumption (Percent)	Weighted Average Percent Lipids
Freshwater fishes	12	4.8
Saltwater fishes	61	2.3
Saltwater molluscs	9	1.2
Saltwater decapods	18	1.2

Using the percentages for consumption and lipids for each of these groups, the weighted average percent lipids is 2.3 for consumed fish and shellfish.

Neither measured steady-state bioconcentration factors (BCF) nor laboratory data (octanol-water partition coefficient) are available for estimation of BCF at this time.

B. Inhalation

No monitoring information is available on the levels of any chloroethers in ambient air.

C. Dermal

Because of the lack of monitoring data, no evaluation of the importance of dermal exposures can be made for the haloethers.

III. Pharmacokinetics

No information has been encountered in the pharmacokinetics of the haloethers under review.

IV. Effects

1. Acute, Subacute, and Chronic

The acute and subacute oral toxicity of various chlorinated phenyl ethers is summarized in Tables 3 and 4. Because of the lack of experimental detail presented in this summary of unpublished data by Hake and Rowe (1963) these results are difficult to interpret. However, the reported results on "highly purified" pentachlorophenyl ether compared to the other pentachlorophenyl ether suggest that impurities may be major toxic constituents.

Hake and Rowe (1963) report that "small amounts" of hexachlorodiphenyl ether may cause acneform dermatitis in man.

2. Joint Action with Other Toxicants

No information is available.

3. Teratogenicity

No information is available.

4. Mutagenicity

No information is available.

5. Carcinogenicity

No information is available.

Table 3. Chlorinated Phenyl Ethers Summary of Single-Dose Oral Feeding Studies on Guinea Pigs (Hake and Rowe, 1963)

	After 4 days		After 30 days	
Total Number of Chlorines	Lethal dose (mg/kg)	Survival dose (mg/kg)	Lethal dose (mg/kg)	Survival dose (mg/kg)
1 x Cl	700	200	600	100
2 x Cl	1,300	400	1,000	50
3 x Cl	2,200	400	1,200	200
4 x Cl	3,000	400	50	0.5
5 x Cl	3,400	1,800	100	5
6 x Cl	3,600	400	50	5

Table 4. Chlorinated Phenyl Ethers: Results of Repeated Oral Feeding of Rabbits (Hake and Rowe, 1963)

Total Number of Chlorines	Dose (mg/kg)	Number of doses*	Number of days	Effect
1	100	19	29	None
2	100	19	29	Mild liver injury
3	100	5	12	Death
	50	20	29	Slight liver injury
	10	20	29	No effect
4	50	4	10	Death
	5	20	29	Severe liver injury
5	50	8	21	Death
	100**	20	29	Moderate liver injury No growth
	10**	20	29	Slight liver injury
	1**	20	29	No effect
6	5	8	10	Death
	1	20	28	Severe liver injury
	0.1	20	28	No effect

*Animals dosed 5 days/week x 4 weeks unless death intervened. Vehicles not specified.

**Highly purified pentachlorophenyl ether.

V. Criterion Rationale

A. Existing Standards

The Occupational Safety and Health Administration (38 FR 23540) has set a time-weighted average value of 500 $\mu g/m^3$ for the following aromatic chloroethers in the air of the working environment: monochlorophenyl phenyl ether, dichlorophenyl phenyl ether, trichlorophenyl phenyl ether, tetrachlorophenyl phenyl ether, and pentachlorophenyl phenyl ether. This value has also been adopted by the American Conference of Governmental and Industrial Hygienists (ACGIH, 1974). The standard is designed to prevent the formation of chloracne in exposed workers.

B. Current Levels of Exposure

As detailed in Section II, only limited information is available on the extent of human exposure to haloethers in water and no information is available on ambient levels of haloethers in air or food. Quantitative estimates of human exposure cannot be made.

C. Special Groups at Risk

Individuals working with haloethers or living in areas where these haloethers are produced are probably at greater risk than the general population.

D. Basis for the Standard

As indicated in Section V.A., the TLV for chlorophenyl phenyl ethers is 500 $\mu g/m^3$. By a process analogous to that used by Stokinger and Woodard (1958), this standard could be used to calculate a water criterion. However, since the TLV for these compounds is based on preventing chloracne, rather than chronic toxicity, such a calculation would not be appropriate.

There are not sufficient toxicologic data to calculate exposure criteria for other haloethers covered in this document.

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